Atomistic-Level Simulation of the Vibration Spectrum of Quantum Dot Crystals

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Quantum dot crystal structures (QDCs) have been recently proposed as the building blocks of novel thermoelectric devices. These structures consist of several vertically stacked layers of regimented arrays of quantum dots (QDs). Three-dimensional periodicity differentiates a QDC structure from a 2-D quantum dot superlattice. If the quantum dots are sufficiently close for the wave functions of the confined electrons to overlap, mini-bands are formed. Electronic transport through mini-bands can be tuned such as to increase the carrier mobility, the Seebeck coefficient compared to bulk properties, and, as a result, will lead to the enhancement of the thermoelectric figure-of-merit. The possibility of changing the sign of the Seebeck coefficient in the material with the same type of doping is an additional benefit of using a QDC for the design of both legs in thermoelectric devices. On the other hand, QDs are scattering centers for lattice vibrations. The lattice thermal conductivity is expected to decrease due to the modification of the acoustical phonon spectrum. The best up-to-date estimations of lattice thermal conductivity are based on volume fraction of quantum dots in QDC. However, this approach is insufficient for high precision modeling and design of novel thermoelectric devices.

We show that adequate modeling of the acoustic phonon spectrum in QDC requires an atomistic approach, which is able to describe the lifting of TA phonon mode degeneracy and the formation of stop-bands (see Fig. 1). The built-in strain and its effect on the phonon spectrum is computed by supplementing the Keating model with anharmonicity corrections. We have found that QDCs can be described as macroscopic objects with effective energy-dependent Grüneisen and biaxial strain coefficients. Our investigation shows that QDCs are very beneficial for creation of thermoelectric devices and are much more sensitive to deformation than bulk constituent materials.

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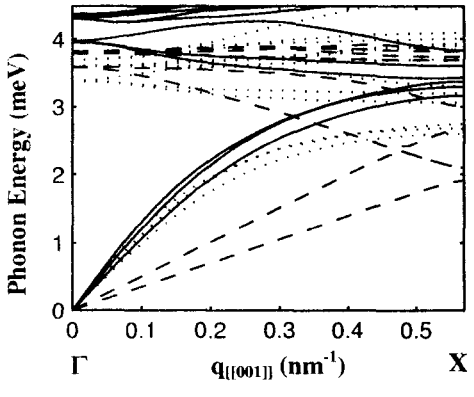

Figure 1: Dispersion in the [[001]] quasi-crystallographic direction, which is along the growth direction of Ge/Si cubical QDC with periodicity $D = 5.5$nm and QD size $L = 2.5$nm. Dashed lines correspond to the continuum approximation; Dotted lines correspond to calculations using the standard Keating model; Solid lines correspond to calculations using the Keating model with anharmonicity corrections.